Carbon Nitride Supported High-Loading Fe Single-Atom Catalyst for Activating of Peroxymonosulfate to Generate 1O2 with 100% Selectivity.

Abstract

Singlet oxygen (1O2) is an excellent active species for the selective degradation of organic pollutions. However, it is difficult to achieve high efficiency and selectivity for the generation of 1O2. In this work, we develop a graphitic carbon nitride supported Fe single-atoms catalyst (Fe1/CN) containing highly uniform Fe-N4 active sites with a high Fe loading of 11.2 wt%. The Fe1/CN achieves generation of 100% 1O2 by activating peroxymonosulfate (PMS), which shows an ultrahigh p-chlorophenol degradation efficiency. Density functional theory calculations results demonstrate that in contrast to Co and Ni single-atom sites, the Fe-N4 sites in Fe1/CN adsorb the terminal O of PMS, which can facilitate the oxidization of PMS to form SO5•-, and thereafter efficiently generate 1O2 with 100% selectivity. In addition, the Fe1/CN exhibits strong resistance to inorganic ions, natural organic matter, and pH value during the degradation of organic pollutants in the presence of PMS. This work develops a novel catalyst for the 100% selective production of 1O2 for highly selective and efficient degradation of pollutants.